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6. AUTHOR(S) Prof. Pei-Wen Li				5d. PROJECT NUMBER		
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Project Title: Innovative Ge Quantum Dot Functional Sensing/Metrology Devices

Period: May 1st 2014–April 30 2017

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Abstract

Through this one-year (2014.05–2015.04) project, we have successfully developed cutting-edge fabrication technologies for the realization of innovative Germanium (Ge) quantum dot (QD) sensing and metrology devices, including (1) QD single-electron transistors (SETs) for single charge detection at temperature as high as 300K, (2) spatial thermometry with a detection temperature as high as 175 K in a spatial resolution less than 10nm and temperature accuracy of sub-kelvin, and (3) coupled-QD (CQD) phototransistors for visible to near IR photodetections.

Motivation to employ QDs for primary sensing and metrology devices is strong in light of the distinctive Coulomb blockade and quantum confinement effects occurring to nanometer-scaled QD structures when their size is scaled down to much less than their corresponding exciton Bohr radius. These peculiar quantum effects give rise to fantastic, size-tunable electronic structures as well as optical and thermoelectric properties for QDs, opening up access to wide-ranging applications in logics, computing, photonics, metrology, and energy conversion devices. A QD SET with narrow, well-spaced energy levels and sharp differential conductance peaks has a Seebeck coefficient independent of material parameters that can be measured in terms of the fundamental electronic charge, making it suitable for metrological standards in various aspects since a QD SET is extremely sensitive to charge number and local temperature with unprecedented precision. Accordingly we have made progresses in the innovative functionalities of QD devices, to enhance improvement of device performance and break through traditional fabrication bottlenecks and classical physical limitation based on a CMOS-compatible technology.

Project Outcome

This project starts from May 01 2014. In the past year, we have made a great success on (1) the growth of “designer” spherical Ge QDs with desired QD size at targeted spatial locations within SiO₂, Si₃N₄, and Si substrate. In addition to the precise control over the addressability, geometrical sizes, and morphological shapes of the Ge QDs, (2) we are also able to do exquisite strain engineering and band gap engineering in our Ge QDs by tailoring the QD size and their embedding within either SiO₂ or Si₃N₄ or even in Si substrate. On the practical side, through the exquisite control of the Ge QD geometries and electronic structures, (3) we have successfully fabricated and previously-impossible heterostructure devices including Ge QD photodiodes and phototransistors for broadband photodetection and amplification as well as Ge QD single electron transistors for Nanothermometry. These QD devices/sensors feature excellent sensitivity on charge number, local temperature, and photoresponsivity in the visible to near IR wavelength.

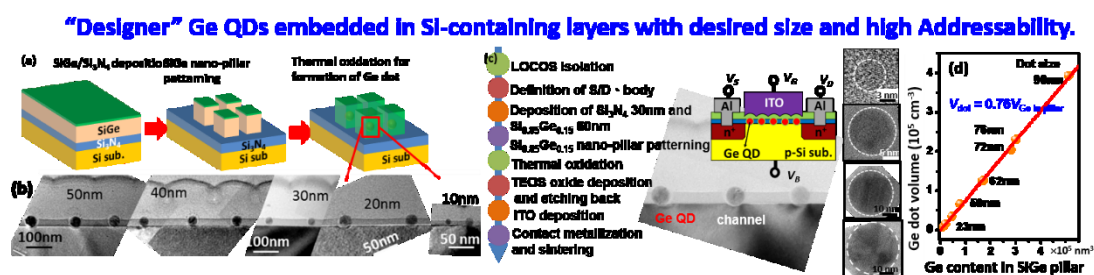
- **“Designer” Ge QDs of desired size with high addressability**

In order to fully exert quantum mechanics effects arising from zero-dimensional quantum-dot structures, precise QD size/shape control with great addressability and contacting with nanoelectrodes is definitely essential for practical application. Detailed knowledge and understanding of how the QDs are created, and especially their interactions with their local environments are therefore crucial to achieve a high level of nanofabrication control on an otherwise random growth process.

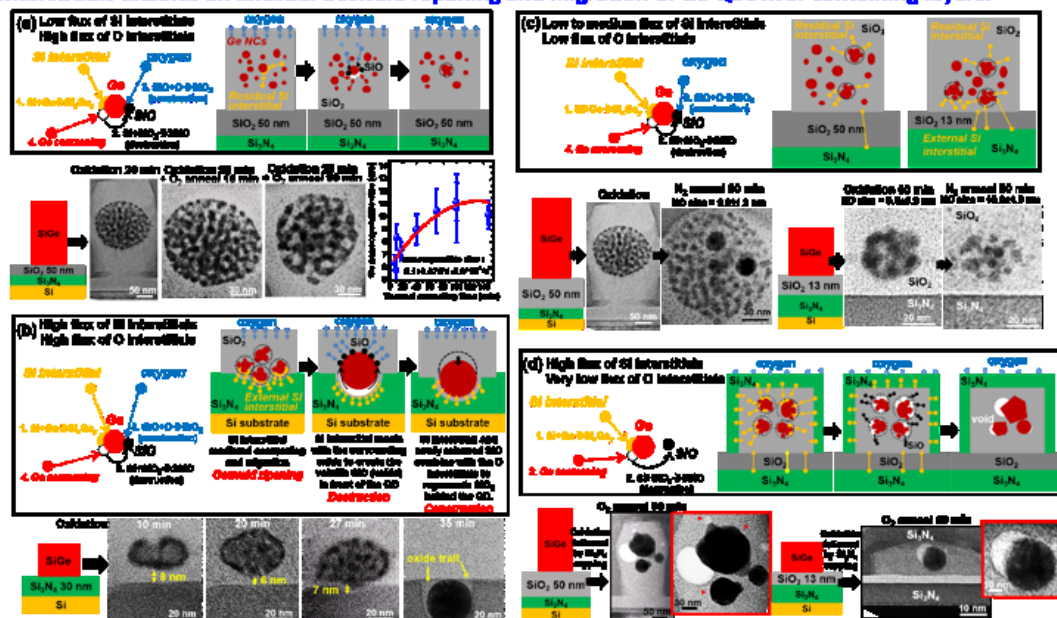
We are fortunate to discover unusual and counter-intuitive interactions of the “Symbiotic” semiconductors Ge/Si, enabling the growth of spherical Ge quantum dots (QDs) of desired diameters at targeted spatial locations with controlled depths of penetration into Si-containing layers such as SiO₂, Si₃N₄, and Si substrate. This was achieved using the exquisite control available through lithographic nanopatterning and selective oxidation of the nanopatterned SiGe layers. We reported a unique, cooperative mechanism that involves the interplay of Ge, Si, and Oxygen interstitials enabling unusual Ostwald Ripening and migration behaviors for Ge nanocrystallites embedded within a SiO₂ matrix and subsequently within Si₃N₄ or Si layers. Our extensive experimentation and characterization of the intriguing anomalous migration behavior and morphology changes of the Ge QDs using Raman Spectroscopy and High Resolution Transmission Electron Microscopy (HRTEM), have led to our proposing mechanisms that are radically different from the accepted paradigms.

Ge nanocrystallites generated by the selective oxidation of SiGe nano-pillars appear to be very sensitive to the presence of Si interstitials generated either from within the oxidized nano-pillars themselves, or from the adjacent Si₃N₄ layers, or

from the Si substrate. With no external source of Si interstitials, very little change in the size and morphology of Ge nanocrystallites is observed during thermal annealing in an H₂O ambient. When both Si and Oxygen interstitials are present in high concentrations, for instance, a high flux of Si interstitials is emitted through catalytic local oxidation of Si₃N₄ layers in proximity to the growing Ge nanocrystallites. Ostwald Ripening occurs concurrently, propelling the Ge nanocrystallite to migrate towards the source of the Si interstitials—the adjacent Si₃N₄ layers. A cooperative phenomenon is observed wherein the Si interstitials aid in both the migration and coarsening of these Ge nanocrystallites into spherically-shaped Ge QDs. In turn, the Ge nanocrystallites appear to enhance the generation of Si interstitials through catalytic decomposition of the Si₃N₄ layers. The Ge nanocrystallite movement occurs by virtue of the fact that the Si interstitials created in front of the nanocrystallites combine with O interstitials to regenerate SiO₂ behind the Ge nanocrystallites on their migration path.



“Cooperative Destruction/Construction” mechanism that involves the interplay of Ge, Si, and Oxygen interstitials enables an unusual Ostwald Ripening and migration of Ge QDs in Si-containing layers.



Yet another, SiO₂-enabled, void formation mechanism is observed for Ge nanocrystallites migrating through SiO₂ layers. This type of migration also ultimately transforms the Ge nanocrystallite clusters into fully coalesced spherical QDs. Here, in

- Matrix-induced Strain Engineering

“Size-tunable” strain engineering in Ge QDs embedded in Si-containing layers. Tensile strains of 0.3–0.7% and compressive strain of 0.6–4% can be generated from SiO₂ and Si₃N₄ layers, respectively.

(a) TEM image of SiGe shell-encapsulated Ge QDs. (b) Depth profile of Si and Ge atomic percentages. (c) SAED pattern with zone axes. (d) HRTEM image of a single Ge QD with SiGe shell. (e) HRTEM images of Ge QDs with different diameters (216 nm, 18 nm, 100 nm, 50 nm, 110 nm) and their corresponding size distribution histograms. (f) Schematic of the strain profile $E_r(r)$ showing tensile strain in the SiGe shell and compressive strain in the Ge QD core.

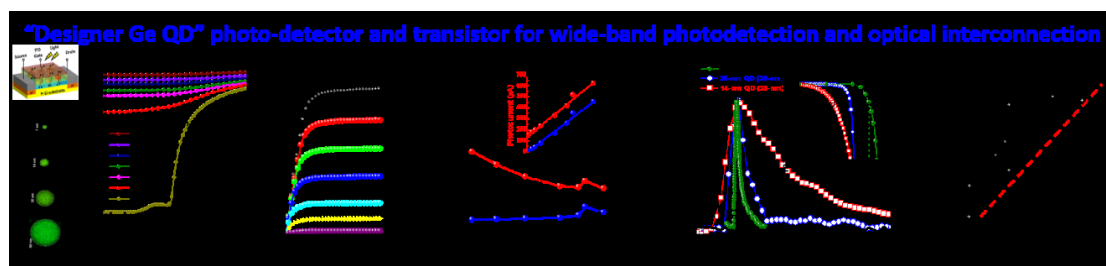
- **Quantum-size-tunable band gap engineering**

On the practical side, through the exquisite control offered by the above

mechanisms, we have successfully fabricated and tested previously-impossible heterostructure devices including Ge QD photodetectors, phototransistors, and single-electron transistors, and QD Coulomb blockade thermometer.

Designer Ge QD photodetectors and phototransistors for Broadband (Near-UV—Near IR) photosensing and amplification

The first experimental example of designer Ge QDs sensing device is the size-tunable Ge QD Metal-Oxide-Semiconductor photodetectors with size tunable Ge QDs for near-ultraviolet to near-infrared photosensing with high figure of merits of low dark current density (1.5×10^{-3} mA/cm²), superior photo current-to-dark current ratio (13,500), high photoresponsivity (2.2 A/W), and fast response time (5 ns), which are ready for direct integration with Si CMOS electronic circuits. Most importantly, the detection wavelength of the Ge QD is tunable from near infrared to near ultraviolet by reducing the QD size from 50 to 7 nm as well as the optimal photoresponsivity is tailored by the Ge QD size and the effective thickness of gate dielectrics. We further exploit this novel approach to construct high performance Ge-QD MOS phototransistor based on this “designer Ge QD MOS structure”. The Ge-QD MOS phototransistors feature extremely low dark current densities ($I_{off} \sim 0.27$ pA/mm²), high I_{on}/I_{off} ($>10^6$), steep subthreshold swing (~ 175 mV/dec at 300 K), superior external quantum efficiency ($>100\%$), and fast response time in nanoseconds under illumination of 850 nm, providing a core building block for high-performance Ge optical transducers for on-chip optical interconnect applications.



A very fast temporal response time of 1.4 ns together with a 3 dB bandwidth of 410 MHz were measured on our Ge-QD MOS phototransistor biased at $V_G = +6V$ and $V_D = +3V$. The measured PL lifetime of the Ge QDs at 20K is approximately 1.7ns. According to the temperature dependency of PL intensities in the range of 20—300K, we estimate that the recombination lifetime at 300K is shorter than 0.2ns (which is beyond the temporal resolution limit of our TRPL system). The deduced photocarrier lifetime is approximately 1/10 of the temporal response time measured on the Ge-QD phototransistors. The measured photocarrier lifetime of 1.7ns at 20K for Ge QDs is three orders longer than the lifetime (~ 1 ps) for oxygen-implanted Ge

thin film, a great indication of enhanced electron-electron interactions. The photo-carrier lifetime as short as sub-nanosecond measured on our Ge QDs suggests the great promise of fast operation of Ge-QD photoconductive switches for on-chip optical interconnect applications.

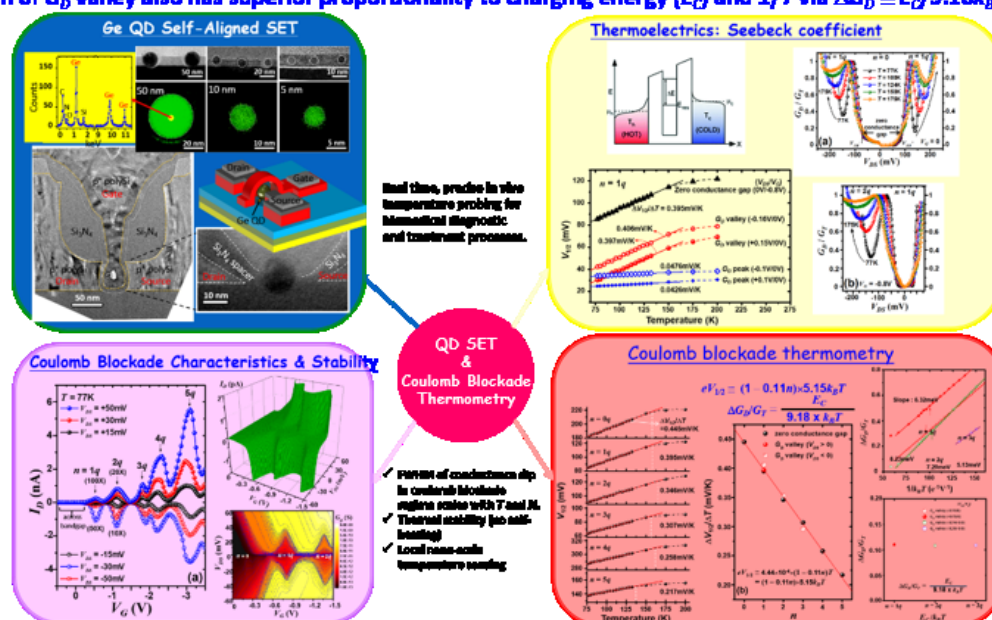
Solid-state Ge Quantum-dot Coulomb-blockade Nanothermometer

The second, very important experimental demonstration based on our designer Ge QD structure is the Ge QD single-hole Nanothermometer. Using a CMOS fabrication approach, we have successfully demonstrated Ge-QD Single-Hole Transistors (SHTs) with self-aligned electrodes based on our exquisite, precise capability to produce a single 10-nm Ge QD that is precisely placed between source and drain electrodes via symmetrical tunnel barriers of $\text{Si}_3\text{N}_4/\text{SiO}_2$. High-performance Ge-QD SHT features distinctive Coulomb-blockade, oscillatory current behaviors with peak-to-valley ratios (PVCRs) in excess of 100 coupled with extremely low leakage ($\sim \text{fA}$) at $T = 77 - 300\text{K}$ thanks to self-aligned gate electrode that effectively suppresses gate-induced tunneling barrier lowering. This experimental demonstration of Ge-QD SHTs offers promising potentials for nanothermometry and other conceivable applications. The semiconductor QD Coulomb-blockade thermometry is based on the extraordinary temperature-dependent oscillatory differential conductance (G_D) characteristics of Ge-QD SHTs in the few-hole regime. Full-voltage width-at-half-minimum, $V_{1/2}$, of G_D valleys exhibits a fair linear dependency on the charge number (n) and temperature within the QD in a relationship of $eV_{1/2} \cong (1-0.11n) \times 5.15k_B T$, providing the primary thermometric quantity. The depth of each G_D valley also possesses good proportionality to its corresponding charging energy (E_C) and $1/T$ via $\Delta G_D \cong E_C/9.18k_B T$, providing another thermometric quantity. This experimental demonstration suggests our Ge-QD SHT providing core building blocks for nanothermometers over a wide temperature range with a detection temperature as high as 175 K in a spatial resolution less than 10nm and temperature accuracy of sub-kelvin.

Relentless miniaturization of transistors for boosting drive current and speed has heightened the importance of localized heating problem and thus, nanoscale-spatially resolved temperature monitoring for local hot spots is central for thermal management and reliability analysis. On the other hand, biology and medicine also demand for nanothermometers since real time, precise in vivo local temperature probing is essential in biomedical premature diagnosis and therapeutic process. Our Ge QD nanothermometer is non-invasive and possesses nanometer-scale spatial resolution and temperature accuracy of sub-kelvin,

providing many appealing applications for nanoelectronics, biomedicines and any other conceivable applications.

Ge QD single hole transistor (SHT) operating in few-hole regime exhibits well-spaced energy levels and sharp differential conductance (G_D) peaks. Full-voltage width-at-half-minimum ($V_{1/2}$) of G_D valleys exhibits good linearity with the charge number (n) within the QD and temperature: $eV_{1/2} \equiv (1-0.11n) \times 5.15k_B T$. The depth of G_D valley also has superior proportionality to charging energy (E_C) and $1/T$ via $\Delta G_D \equiv E_C/9.18k_B T$.



Our Ge-QD SHT provides effective building blocks for QD nanothermometers with detection temperature as high as 155 K in a spatial resolution less than 10nm and temperature accuracy of sub-kelvin.

We have reported these fascinating phenomena and mechanisms for Designer Ge QDs embedding within Si-containing layers and their applications for SETs, phototransistors, and Nanothermometry in Nanoscale, Applied Physics Letters, Optical Letters, Physical Chemistry Chemical Physics, Nanoscale Research Letters as follows. There are 8 journal papers, international conference papers published, and 5 invited talks in international conferences, and one best paper award in 2014 IEDMS.

1. **Published Journal Papers** Published papers (Journal name, title, date):

- (1) M. H. Kuo, W. T. Lai, S. W. Lee, and **P. W. Li**, 2015, "High-quality thin-film-like multifold Ge/Si/Ge composite quantum-dot heterostructures for visible to near-infrared photodetection" to appear in Optical Letters .
- (2) O. Korotchenkov, A. Nadtochiy, V. Kuryliuk, Chin-Chi Wang, and **Pei-Wen Li**, 2015 "A model for predicting the thermal conductivity of SiO₂/Ge nanoparticle composites," to appear in Physical Chemistry Chemical Physics.
- (3) M. H. Kuo, W. T. Lai, T. M. Hsu, and **P. W. Li**, 2015, "Designer germanium quantum dot phototransistor for near infrared optical detection and amplification," Nanotechnology, vol. 26, 055203.

- (4) P.-H. Liao, T.-C. Hsu, K.-H. Chen, T.-H. Cheng, T.-M. Hsu, C.-C. Wang, T. George, and **P. W. Li**, "Size-tunable strain engineering in Ge nanocrystals embedded within SiO₂ and Si₃N₄," Appl. Phys. Letts. vol. 105, 172106 (2014).
- (5) K. H. Chen, C. C. Wang, Tom George, and **P. W. Li**, "The pivotal role of SiO for formation in the migration and Ostwald Ripening of Ge quantum dots," Appl. Phys. Lett., vol. 105, 122102 (2014).
- (6) Chung-Yen Chien, Yu-Jui Chang, Ching-Chi Wang, Ming-Hao Kuo, Wei-Ting Lai, and **Pei-Wen Li**, 2014, "Size tunable Ge quantum dot metal-oxide-semiconductor photodiodes with low dark current and high responsivity for near ultraviolet to visible applications," Nanoscale, 6 (10), 5303 – 5308
- (7) K. H. Chen, C. C. Wang, T. George, and **P. W. Li**, "The role of Si interstitials in the migration and growth of Ge nanocrystallites under thermal annealing in an oxidizing ambient", Nanoscale Research Letter, vol. 9, 339 (2014).
- (8) I. H. Chen, W. T. Lai, and **P. W. Li**, 2014, "Designer Ge quantum dot Coulomb blockade thermometry,"Appl. Phys. Lett., vol. 104, 243506.
- (9) Chung-Yen Chien, Yu-Jui Chang, Ching-Chi Wang, Ming-Hao Kuo, Wei-Ting Lai, and **Pei-Wen Li**, 2014, "Size tunable Ge quantum dot metal-oxide-semiconductor photodiodes with low dark current and high responsivity for near ultraviolet to visible applications," Nanoscale, 6 (10), 5303 – 5308.
- (10) O. Korotchenkov, A. Nadtochiy, V. Kuryliuk, Chin-Chi Wang, **Pei-Wen Li**, A. Cantarero, 2014, "Thermoelectric energy conversion in layered structures with strained Ge quantum dots grown on Si surfaces," European Physical Journal B, vol. 87, 64.

2. Paper currently under review (journal name, title, date accepted):

- (1) W. T. Lai, K. H. Chen, T. George, and P. W. Li, "Size-tunable Ge quantum dot gate/SiO₂/SiGe channel for self-aligned Insta metal-oxide-semiconductor devices," submitted to Nanoscale.
- (2) W. T. Lai, K. C. Yang, T. C. Hsu, P. H. Liao, T. George, and P. W. Li, "A novel approach to generate self-aligned SiO₂/Ge/SiO₂/SiGe gate-stacking heterostructures in a single fabrication step," submitted to Appl. Phys. Lett.

3. Conference paper/poster/presentation (conf. name, title, date):

- (1) Ming-Hao Kuo, Chung-Yen Chien, Po-Hsiang Liao, Wei-Ting Lai, and Pei-Wen Li, "Size tunable Ge quantum dot phototransistors for optical interconnects with high figure of merits", in VLSI-TSA, Hsinchu, Taiwan, 27- 28 April, 2015.
- (2) P. W. Li and Wei-Ting Lai, "A novel approach to generate self-aligned SiO₂/Ge/SiO₂/Ge gate-stacking heterostructures in a single fabrication step", in 2015 Material Research Society Spring Meeting, San Francisco, USA, 6 - 10 April, 2015.

- (3) Shuo-Huang Yuan , Zing-Way Pei , Pei-Wen Li , Yi-Jen Chan , "High responsivity pentacene phototransistor achieved by surface plasmon enhancement," 2014 MRS Fall Meeting, Boston, Massachusetts, USA (2014/11/30~12/5)
- (4) Ching-Chi Wang, Yi-Yeh Hsiao, Inn-Hao Chen, Tom George, and Pei-Wen Li, "Geometry-dependent phases and electrical properties in nickel silicide nanowires ," 2014 International Electron Devices and Materials Symposium (IEDMS), Haulien, Taiwan. (Nov19-21, 2014)
- (5) Jyun-Yi Jhong , Pei-Wen Li, Tien-Chun Lee, Geng-Tai Ho, and Shih-Kuei Ma , "Device scaling design of 700V super-junction MOSFETs for high figure of merits," 2014 International Electron Devices and Materials Symposium (IEDMS), Haulien, Taiwan. (Nov19-21, 2014)
- (6) Kuan-Hung Chen, Ching-Chi Wang, Tom George and Pei-Wen Li, "The role of Si interstitials in the migration and growth of Ge nanocrystallites under thermal annealing in an oxidizing ambient," 2014 International Electron Devices and Materials Symposium (IEDMS), Taiwan. (Nov19-21, 2014)
- (7) Ming-Hao Kuo, Wei-Ting Lai, Tzu-Min Hsu, and Pei-Wen Li, "Designer Germanium Quantum Dot Phototransistor for Near Infrared Optical Detection and Amplification," 2014 International Electron Devices and Materials Symposium (IEDMS), Haulien, Taiwan. (Nov19-21, 2014)
- (8) P. H. Liao, T. C. Hsu¹, K. H. Chen, T. H. Cheng, T. M. Hsu, and P. W. Li, "Size Tunable Strain for Germanium Quantum Dots Embedded within SiO₂ and Si₃N₄," 2014 International Electron Devices and Materials Symposium (IEDMS), Haulien, Taiwan. (Nov19-21, 2014)
- (9) Wei-Ting Lai, Kuo-Ching Yang, Ting-Chia Hsu, Po-Hsiang Liao, Thomas George, and Pei-Wen Li, "A Self-aligned Ge/SiO₂/Si_{0.5}Ge_{0.5} gate-stacking heterostructure generated in a single-step fabrication," 2014 International Electron Devices and Materials Symposium (IEDMS), Haulien, Taiwan. (Nov19-21, 2014)
- (10) M. H. Kuo, W. T. Lai, H. T. Chang, S. W. Lee, and P. W. Li., "High-quality thin-film-like multifold Ge/Si/Ge composite quantum-dot hetero-structures for visible to near-infrared photodetection," 2014 International Conference on Solid-State Devices and Materials, Tsukuba, Japan (Sep. 8-11, 2014).
- (11) W. T. Lai, K. C. Yang, T. C. Hsu, P. H. Liao, T. George, and P. W. Li, "A self-aligned Ge/SiO₂/Si_{0.4}Ge_{0.6} gate-stacking heterostructure generated in a single fabrication step," 2014 International Conference on Solid-State Devices and Materials, Tsukuba, Japan (Sep. 8-11, 2014).
- (12) Wei-Ting Lai, Kuo-Ching Yang, Ting-Chia Hsu, Po-Hsiang Liao, Thomas George, and Pei-Wen Li, 2014 "A Novel Approach to Generate Self-aligned Ge/SiO₂/SiGe Gate-stacking Structures in a Single Fabrication Step," 2014 Silicon Nanoelectronics

Workshop. The Workshop, Hawaii, USA 2014/6/8~2014/6/9.

- (13) Ming-Hao Kuo, Wei-Ting Lai, Tzyy-Min Hsu, and Pei-Wen Li 2014, "Designer Germanium Quantum Dot Phototransistor for Near Infrared Optical Detection and Amplification," 2014 Silicon Nanoelectronics Workshop. The Workshop, Hawaii, USA 2014/6/8~2014/6/9.
- (14) Po-Hsiang Liao, Ting-Chia Hsu, Tzu-Hsuan Cheng, Tzu-Min Hsu, and Pei-Wen Li, 2014, "Size Tunable Strain and Interfacial Engineering of Germanium Quantum Dots," 2014 Silicon Nanoelectronics Workshop. The Workshop, Hawaii, USA 2014/6/8~2014/6/9.
- (15) Wei-Ting Lai, Kuo-Ching Yang, Ting-Chia Hsu, Po-Hsiang Liao, Thomas George, and Pei-Wen Li" A Novel Approach to Generate Self-aligned Ge/SiO₂/SiGe Gate-stacking Structures in a Single Fabrication Step," 2014 Silicon Nanoelectronics Workshop. The Workshop, Hawaii, USA 2014/6/8~2014/6/9.
- (16) I. H. Chen, C. C. Wang, and P. W. Li, 2014, "Designer Ge quantum dots Coulomb blockade thermometry," oral presentation in 2014, VLSI-TSA, HsinChu, April 28 (2014).

4. **Invited talks**

- (1) [P. W. Li, \(Invited Talk\)](#) 2014, "Size tunable Ge quantum dot phototransistors for optical interconnects with high figure of merits," Photonics 2014: 12th International Conference on Fiber Optics and Photonics, Kharagpur, India, 13 - 16 December, 2014.
- (2) [P. W. Li, \(Invited Talk\)](#) 2014, "Designer germanium quantum dots for functional sensing/metrology devices," 2014 EITA-New Materials, Tainan, Taiwan, Nov. 22-23 2014.
- (3) [P. W. Li, \(Invited Talk\)](#) 2014, "Designer Germanium Quantum-Dot for Nanoelectronics and Nanophotonics Devices," 2014 International Electron Devices and Materials Symposium (IEDMS), Hualien, Taiwan. (Nov19-21, 2014)
- (4) [P. W. Li, \(Invited Talk\)](#) 2014, "Designer Ge quantum dot single electron transistor and Coulomb blockade thermometry", The 6th IEEE International Nanoelectronics Conference, IEEE INEC 2014, 2014/7/28~7/31 Sapporo, Hokkaido, Japan.
- (5) [P. W. Li, \(Invited talk\), 2014](#), "The Curious case of Germanium Quantum dots: Fantasy migration and Ripening behavior of Ge under Si interstitials oxidation," IUMRS-ICEM, Taipei, June 11-June 14 (2014).

5. Award for best paper, best poster (title, date):

- (1) Best Paper in 2014 International Electron Devices and Materials Symposium (IEDMS), Hualien, Taiwan. (Nov19-21, 2014), Ming-Hao Kuo, Wei-Ting Lai, Tzu-Min Hsu, and Pei-Wen Li, "Designer Germanium quantum dot phototransistor for near infrared optical detection and amplification".

6. Award of fund received related to your research efforts (name, amount, date):

- (1) "Innovative Ge Quantum Dot Functional Sensing/Metrology Devices" from Ministry of Science and Technology, MOST 102-2221-E-009-195-MY3, NT\$6,312,000. (2014/05 – 2017/04)
- (2) "Design and Fabrication of 600V Super-junction Power MOSFET ," Joint Development project with Episil Technology, from HsinChu Science-based Industrial Park, 103A04, NT\$1,300,000. (2014. 05–2015.04)
- (3) "Innovative Ge quantum Dot Functional Sensing/Metrology Devices" from Asian Office of Aerospace R&D, FA2386-14-1-4008, NT\$1,200,000.

7. IP disclosure/Patent (title, date submitted):

- (1) "Method for manufacturing gate stack structure in insta-metal-oxide-semiconductor Field-effect-transistor", by Wei-Ting Lai, T. George, and P. W. Li, Taiwan and US patents, (Pending), Dec. 2014.
- (2) "Method For Forming a Thermoelectric Film Having a Micro Groove," Sheng-Wei Lee, Yi-Fan Niu, Pei-Wen Li, Cheng-Lun Hsin, Chung-Jen Tseng, Taiwan and US patents, (Pending), July. 2014.
- (3) "Manufacturing Method for Forming Substrate with Silicon-Germanium Epitaxial Layer", Sheng-Wei Lee, Hung-Tai Chang, Kan-Rong, Lee, Pei-Wen Li, Cheng-Lun Hsin, Taiwan and US patents, (Pending), Jan. 2014.

8. Visited AFRL/DoD installation with AOARD WoS program (Location, date):

Dayton Ohio, April 6